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THEORETICAL CONSIDERATIONS REGARDING ACTIVE SAMPLING OF TIME-DEPENDENT RADON (THORON) ATMOSPHERES

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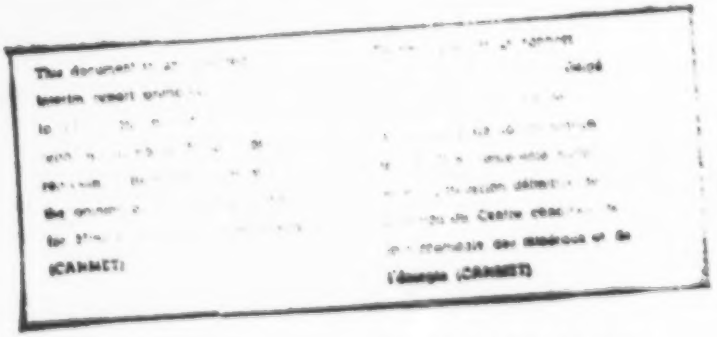

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THEORETICAL CONSIDERATIONS REGARDING ACTIVE SAMPLING OF TIME-DEPENDENT RADON (THORON) ATMOSPHERES

by

J. Bigu*

ABSTRACT

The theory of active sampling of the short-lived decay products of radon and thoron under time-dependent conditions is discussed. Active sampling, i.e., sampling through an 'absolute' filter by means of a sampling pump, is investigated under conditions in which the airborne concentration of the decay products of radon and thoron varies with time in a predictable fashion. Analytical expressions have been derived which describe the dynamic behaviour of sampling under sinusoidal, linear, exponential and step-wise variations in radon and thoron progeny atmospheres. The study presented here includes expressions for the calculation of the growth and/or decay of activity, differential and integrated, during the sampling period and after sampling. This work has practical applications to personal dosimetry and environmental monitoring in residential areas and at working sites in uranium-related industries.

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OBSERVATIONS THÉORIQUES SUR L'ÉCHANTILLONNAGE ACTIF DANS L'ATMOSPHÈRE DU RADON (THORON) EN FONCTION DE TEMPS

par

J. Bigu*

RÉSUMÉ

Ce rapport présente une discussion sur la théorie de l'échantillonnage actif des produits de désintégration à vie courte du radon et du thoron dans des conditions en fonction de temps. L'échantillonnage actif, c.-à-d. l'échantillonnage à travers un filtre absolu à l'aide d'une pompe d'échantillonnage, a été étudié dans des conditions où la concentration des produits de désintégration du radon et du thoron en suspension dans l'air variait avec le temps de manière prévisible. On en a tiré des expressions analytiques qui décrivent le comportement dynamique de l'échantillonnage lors des variations sinusoïdales, linéaires, exponentielles et étagées des produits de filiation du radon et du thoron dans l'atmosphère. L'étude présentée dans ce rapport comprend les expressions servant à calculer le grandissement et la désintégration de l'activité différentielle et intégrée au cours et après la période d'échantillonnage. Enfin, ce travail a des applications pratiques pouvant servir à la dosimétrie personnelle et au monitorage environnemental des quartiers résidentiels et du milieu de travail dans les industries ayant rapport à l'uranium.

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INTRODUCTION

Monitoring of radiation conditions at working sites is important to assess radiological hazards to occupational workers, and for air quality control purposes and engineering applications. A variety of analytical techniques, methods, and instrumentation have been developed over the past few years. Except for isolated cases, however, these analytical techniques and methods are based on the assumption that radiation conditions remain constant during the sampling period (1-9). This assumption is far from realistic, as radiation conditions in the field are highly dependent on a number of factors, including meteorological conditions. Hence, radiation conditions rarely remain constant over extended periods.

Although, to a large extent, radiation conditions in the field are only partly predictable, it is of great practical interest to derive analytical techniques, and to study the response, behaviour and performance of radiation instrumentation, under well-defined non steady-state radiation conditions. Studies of this nature provide the background for correct interpretation of field data.

This paper presents the theory of active sampling of the short-lived decay products of radon and thoron, i.e., progeny, in time-dependent radioactive atmospheres, in which the radiation levels of the above radioisotopes vary with time in a predictable fashion. Analytical expressions are derived which describe the dynamic response of active sampling under sinusoidal, linear, exponential, and step-wise variations in radon (thoron) progeny atmospheres. The study presented here includes the calculation of: the growth of activity versus time, i.e., differential activity during the sampling period; the decay of activity versus time after the sampling period; the integrated activity during the sampling period, or part thereof; and the integrated activity after the

sampling period, or part thereof. It should be noted that active sampling is to be understood as sampling of an atmosphere through a filter by means of a pump. Furthermore, the reader should be aware that decay products, daughter products, short-lived decay products and progeny are equivalent in the context of this paper and these terms will be used here interchangeably.

The present study is relevant for personal dosimetry and environmental monitoring of the radon and thoron progeny in residential areas (e.g., houses), uranium mines, mills, and other uranium-related industries. The expressions given here have been used extensively elsewhere by the author (10).

THEORETICAL BACKGROUND

The rate of growth of activity on the filter during sampling is given by the following expression:

$$\frac{dA_m}{dt} = QC_m(t) + \lambda_m A_{m-1} - \lambda_m A_m \qquad \text{Eq 1}$$

where, A_m and A_{m-1} are the activities of the m-th and (m-1)-th member of the radioactive decay chain (pCi or Bq), respectively. $C_m(t)$ is the time-dependent atmospheric concentration of the m-th member of the radioactive chain (pCi/m^3 or Bq/m^3); λ_m is the radioactive decay constant of the m-th member of the radioactive decay chain (s^{-1}); and Q is the sampling flow rate (m^3/s).

The activities on the filter, during the sampling period, at any time (T_s) are obtained by integrating Equation 1 for each radionuclide of interest subject to the initial boundary conditions $A_m = 0$ at $t = 0$, i.e., no activity present before the sampling period.

Expressions for the activity on the filter at time T_w after sampling, i.e., waiting period, can easily be obtained by setting $Q = 0$ in Equation 1, and integrating the remaining expression subject to the boundary condition: $A_m(T_w = 0) = A_m(T_s)$ where T_s is, as indicated above, the sampling

time.

The total activity accumulated in the filter during a sampling time T_{s1} to T_{s2} ($T_{s2} > T_{s1}$) is obtained by integrating $A_m(T_s)$. Thus:

$$IA_mS = \int_{T_{s1}}^{T_{s2}} A_m(T_s)dT_s \qquad \text{Eq 2}$$

where the symbol IAS stands for integrated activity during sampling. Similarly, the total activity during the waiting period T_{w1} to T_{w2} ($T_{w2} > T_{w1}$) is given by:

$$IA_mW = \int_{T_{w1}}^{T_{w2}} A_m(T_w)dT_w \qquad \text{Eq 3}$$

where the symbol IAW stands for integrated activity during the waiting period after the end of sampling.

The solutions of Equation 1 for several forms of the time-dependent concentration $C_m(t)$ have been investigated. The following forms of $C_m(t)$ are discussed:

$$C_m(t) = C_{om}\ (1 + \gamma \sin\omega t) \qquad \text{Eq 4}$$

$$C_m(t) = C_{om}\ (1 + \eta t) \qquad \text{Eq 5}$$

$$C_m(t) = C_{om}\ e^{\delta t} \qquad \text{Eq 6}$$

$$\left.\begin{aligned} C_m(t) &= q\ C_{om} \\ \text{where } q &= 1 \text{ for } t \leq t_o, \text{ and} \\ q &\neq 1 \text{ but } q \gtrless 0 \text{ for } t > t_o \end{aligned}\right\} \qquad \text{Eq 7}$$

It should be noted that for the initial boundary condition, i.e., $t = 0$, Equations 4 to 7 reduce to C_{om}, where the index o is used to indicate the initial, steady-state, condition. The symbols γ, η, δ and q are perturbation amplitudes pertaining to Equations 4 to 7, respectively. It should also be noted that for the case $\gamma = 0$, $\eta = 0$, and $\delta = 0$, Equations 4 to 6 reduce to the initial steady-state condition.

Equations 4 to 7 can be written in a more condensed form. Thus:

$$C_m(t) = C_{om}\, F(t) \qquad \text{Eq 8}$$

The function F(t) can be thought of as a perturbation introduced to the otherwise undisturbed, steady-state, condition.

Notice that for small values of δ, Equations 5 and 6, for $\eta = \delta$, are identical, as :

$$e^{\eta t} \cong 1 + \eta t \text{ for } \eta t << 1$$

Large values for δ are only of theoretical interest, but of little practical interest as these rarely reflect actual field conditions. Hence, Equation 6 could be used instead of Equation 5 to solve Equation 1 in most cases of interest, with considerable simplification in the analytical procedure.

The analysis of active sampling under steady-state conditions has been dealt with by the author and other authors elsewhere (1-9), and will not be presented here. The analysis of active sampling in time-dependent radiation conditions is considerably more complex than that under steady-state conditions. Active sampling under time-dependent conditions has been simplified here by making the following assumptions:

a) the sampling flow-rate remains constant during the sampling period;

b) the atmospheric radon and thoron progeny concentrations are in phase. This is not strictly true as indicated by the author elsewhere (11).

c) the perturbation amplitude for all the decay products increases (decreases) by the same amount.

If the indices i, j and k are used to indicate, respectively, RaA or ThA, RaB or ThB and RaC or ThC, Equation 1 can be written for each radionuclide of interest as follows:

$$\frac{dA_i}{dt} = QC_i(t) + \lambda_i A_{i-1} - \lambda_i A_i$$

$$= QC_i(t) - \lambda_i A_i \qquad \text{Eq 9}$$

as $A_{i-1} = 0$ (i.e., i-1 refers to the parent of RaA or ThA, which is a gas and hence is not collected on the filter)

$$\frac{dA_j}{dt} = QC_j(t) + \lambda_j A_i - \lambda_j A_j \qquad \text{Eq 10}$$

and

$$\frac{dA_k}{dt} = QC_k(t) + \lambda_k A_j - \lambda_k A_k \qquad \text{Eq 11}$$

It should be noted that, as the solution of Equation 9, i.e., A_i is needed to solve Equation 10 and so on, the analytical procedure grows considerably in complexity as analysis proceeds from Equation 9 to Equation 11.

Specific solutions for the time-dependent cases expressed by Equation 8 are given below.

SINUSOIDAL CASE

1. Activities During the Sampling Period

For the sinusoidal case, the activities during sampling are given by:

$$A_i(T_s) = A_i^o(T_s) + \frac{QC_{oi}\gamma}{\lambda_i^2+\omega^2}\left[(\lambda_i \sin\omega T_s - \omega\cos\omega T_s) + \omega e^{-\lambda_i T_s}\right] \qquad \text{Eq 12}$$

where, in Equation 12, $A_i^o(T_s)$ is given by:

$$A_i^o(T_s) = \frac{QC_{oi}}{\lambda_i}(1 - e^{-\lambda_i \tau_s}) \qquad \text{Eq 13}$$

$$\begin{aligned} A_j(T_s) = {} & A_j^o(T_s) \\ & + \frac{QC_{oj}\gamma}{\omega^2+\lambda_j^2}\left[(\lambda_j \sin\omega T_s - \omega\cos\omega T_s) + \omega e^{-\lambda_j T_s}\right] \\ & + \frac{QC_{oi}\gamma\lambda_j}{\lambda_i^2+\omega^2}\left[\frac{1}{\omega^2+\lambda_j^2}\left\{(\lambda_i\lambda_j - \omega^2)\sin\omega T_s \right.\right. \\ & \left.\left. - \omega(\lambda_i+\lambda_j)\cos\omega T_s + \omega(\lambda_i+\lambda_j)e^{-\lambda_j T_s}\right\} \right. \\ & \left. + \frac{\omega}{\lambda_j-\lambda_i}(e^{-\lambda_i T_s} - e^{-\lambda_j T_s})\right] \end{aligned} \qquad \text{Eq 14}$$

where

$$A_j^o(T_s) = \frac{QC_{oj}}{\lambda_j}\,(1 - e^{-\lambda_j T_s}) + QC_{oi}\lambda_j\left[\frac{1-e^{-\lambda_i T_s}}{\lambda_i(\lambda_j-\lambda_i)} + \frac{1-e^{-\lambda_j T_s}}{\lambda_j(\lambda_i-\lambda_j)}\right] \qquad \text{Eq 15}$$

and

$$\begin{aligned} A_k(T_s) = A_k^o(T_s) &+ \frac{QC_{ok}\gamma}{\omega^2+\lambda_k^2}\,(\lambda_k \sin\omega T_s - \omega\cos\omega T_s + \omega e^{-\lambda_k T_s}) \\ &+ \frac{QC_{oj}\gamma\lambda_k}{\omega^2+\lambda_j^2}\left[\frac{\lambda_j}{\omega^2+\lambda_k^2}\,(\lambda_k \sin\omega T_s - \omega\cos\omega T_s + \omega e^{-\lambda_k T_s})\right. \\ &\quad + \frac{\omega}{\omega^2+\lambda_k^2}\,(-\lambda_k \cos\omega T_s - \omega\sin\omega T_s + \lambda_k e^{-\lambda_k T_s}) \\ &\quad \left. + \frac{\omega}{\lambda_k-\lambda_j}\,(e^{-\lambda_j T_s} - e^{-\lambda_k T_s})\right] \\ &+ \frac{QC_{oi}\gamma\lambda_j\lambda_k}{\omega^2+\lambda_i^2}\left[\frac{1}{\omega^2+\lambda_j^2}\left\{\frac{\lambda_i\lambda_j-\omega^2}{\omega^2+\lambda_k^2}\,(\lambda_k \sin\omega T_s - \omega\cos\omega T_s + \omega e^{-\lambda_k T_s})\right.\right. \\ &\quad - \frac{\omega(\lambda_i+\lambda_j)}{\omega^2+\lambda_k^2}\,(\lambda_k \cos\omega T_s + \omega\sin\omega T_s - \lambda_k e^{-\lambda_k T_s}) \\ &\quad \left.\left. + \frac{\omega(\lambda_i+\lambda_j)}{\lambda_k-\lambda_j}\,(e^{-\lambda_j T_s} - e^{-\lambda_k T_s})\right\}\right. \\ &\quad + \frac{\omega}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)}\,(e^{-\lambda_i T_s} - e^{-\lambda_k T_s}) \\ &\quad \left. + \frac{\omega}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_j)}\,(e^{-\lambda_k T_s} - e^{-\lambda_j T_s})\right] \end{aligned} \qquad \text{Eq 16}$$

where

$$A_k^o(T_s) = \frac{QC_{ok}}{\lambda_k}\,(1 - e^{-\lambda_k T_s}) + QC_{oj}\lambda_k\left[\frac{1-e^{-\lambda_j T_s}}{\lambda_j(\lambda_k-\lambda_j)} + \frac{1-e^{-\lambda_k T_s}}{\lambda_k(\lambda_j-\lambda_k)}\right]$$

$$+ QC_{oi}\lambda_j\lambda_k\left[\frac{1-e^{-\lambda_i T_s}}{\lambda_i(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{1-e^{-\lambda_j T_s}}{\lambda_j(\lambda_i-\lambda_j)(\lambda_k-\lambda_j)} + \frac{1-e^{-\lambda_k T_s}}{\lambda_k(\lambda_i-\lambda_k)(\lambda_j-\lambda_k)}\right] \qquad \text{Eq 17}$$

where, in Equations 12, 14 and 16, $A_i^o(T_s)$, $A_j^o(T_s)$ and $A_k^o(T_s)$ given, respectively, by Equations 13, 15 and 17, represent the unperturbed, i.e., 'steady-state' or 'DC' contribution corresponding to the radioisotopes i, j and k.

2. Integrated Activities During the Sampling Period

The integrated activities during the sampling period are given by:

$$IA_iS = IA_i^oS + \frac{QC_{oi}\gamma}{\omega^2+\lambda_i^2}\left[\lambda_i F(\sin) - \omega F(\cos) + \omega G_i\right] \qquad \text{Eq 18}$$

where $IA_i^oS = QC_{oi}F_i/\lambda_i$ Eq 19

$$IA_jS = IA_j^oS + \frac{QC_{oj}\gamma}{\omega^2+\lambda_j^2}\left[\lambda_j F(\sin) - \omega F(\cos) + \omega G_j\right] + \frac{QC_{oi}\gamma\lambda_j}{\omega^2+\lambda_i^2}\left[\frac{1}{\omega^2+\lambda_j^2}\left\{(\lambda_i\lambda_j - \omega^2)\,F(\sin\omega) - \omega(\lambda_i+\lambda_j)\,F(\cos) + \omega(\lambda_i+\lambda_j)\,G_j\right\} + \frac{\omega F_{ij}}{\lambda_j-\lambda_i}\right] \qquad \text{Eq 20}$$

where, $IA_j^oS = \frac{QC_{oj}F_j}{\lambda_j} + QC_{oi}\lambda_j\left[\frac{F_i}{\lambda_i(\lambda_j-\lambda_i)} + \frac{F_j}{\lambda_j(\lambda_i-\lambda_j)}\right]$ Eq 21

$$IA_kS = IA_k^oS + \frac{QC_{ok}\gamma}{\omega^2+\lambda_k^2}\left(\lambda_k\,F(\sin) - \omega F(\cos) + \omega G_k\right) + \frac{QC_{oj}\gamma\lambda_k}{\omega^2+\lambda_j^2}\left[\frac{\lambda_j}{\omega^2+\lambda_k^2}\left(\lambda_k F(\sin) - \omega F(\cos) + \omega G_k\right) + \frac{\omega}{\omega^2+\lambda_k^2}\left(-\lambda_k F(\cos) - \omega F(\sin) + \lambda_k G_k\right) + \frac{\omega F_{jk}}{\lambda_k-\lambda_j}\right] + \frac{QC_{oi}\gamma\lambda_j\lambda_k}{\omega^2+\lambda_i^2}\left[\frac{1}{\omega^2+\lambda_j^2}\left\{\frac{\lambda_i\lambda_j-\omega^2}{\omega^2+\lambda_k^2}\left(\lambda_k F(\sin) - \omega F(\cos) + \omega G_k\right)\right.\right.$$

$$- \frac{\omega(\lambda_i+\lambda_j)}{\omega^2+\lambda_k^2} (\lambda_k F(\cos) + \omega F(\sin) - \lambda_k G_k)$$

$$+ \frac{\omega(\lambda_i+\lambda_j)F_{jk}}{\lambda_k-\lambda_j} \Big\} + \frac{\omega F_{ik}}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{\omega F_{kj}}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_j)} \Big] \quad \text{Eq 22}$$

$$IA_k^o S = \frac{QC_{ok}F_k}{\lambda_k} + QC_{oj}\lambda_k \left[\frac{F_j}{\lambda_j(\lambda_k-\lambda_j)} + \frac{F_k}{\lambda_k(\lambda_j-\lambda_k)} \right]$$

$$+ QC_{oi}\lambda_j\lambda_k \left[\frac{F_i}{\lambda_i(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{F_j}{\lambda_j(\lambda_i-\lambda_j)(\lambda_k-\lambda_j)} + \frac{F_k}{\lambda_k(\lambda_i-\lambda_k)(\lambda_j-\lambda_k)} \right] \quad \text{Eq 23}$$

and where,

$$F_i = (T_{s2} - T_{s1}) + \frac{1}{\lambda_i} (e^{-\lambda_i T_{s2}} - e^{-\lambda_i T_{s1}}) ,$$

$$F_j = (T_{s2} - T_{s1}) + \frac{1}{\lambda_j} (e^{-\lambda_j T_{s2}} - e^{-\lambda_j T_{s1}}) ,$$

$$F_k = (T_{s2} - T_{s1}) + \frac{1}{\lambda_k} (e^{-\lambda_k T_{s2}} - e^{-\lambda_k T_{s1}}) ,$$

$F_{ij} = F_j - F_i$; $F_{jk} = F_k - F_j$; $F_{ik} = F_k - F_i$; and $F_{kj} = - F_{jk}$,

$$F(\sin) = (1/\omega)(\cos\omega T_{s1} - \cos\omega T_{s2}) ,$$

$$F(\cos) = (1/\omega)(\sin\omega T_{s2} - \sin\omega T_{s1}) ,$$

$$G_i = \frac{1}{\lambda_i} (e^{-\lambda_i T_{s1}} - e^{-\lambda_i T_{s2}}) ,$$

$$G_j = \frac{1}{\lambda_j} (e^{-\lambda_j T_{s1}} - e^{-\lambda_j T_{s2}}) , \text{ and}$$

$$G_k = \frac{1}{\lambda_k} (e^{-\lambda_k T_{s1}} - e^{-\lambda_k T_{s2}}) .$$

If the total integrated activity is of interest instead of the integrated activity within a sampling interval $T_{s1} \rightarrow T_{s2}$, the sampling times assume the following values $T_{s1} = 0$ and $T_{s2} = T_s$, i.e., the total sampling time.

It should be noted that $IA_i^o S$, $IA_j^o S$ and $IA_k^o S$ in Equations 18, 20 and 22, given by Equations 19, 21 and 23, represent the unperturbed integrated activities corresponding to the radionuclides i, j and k. Furthermore, $A_i^o(T_s)$, $A_j^o(T_s)$ and

$A_k^o(T_s)$, and IA_i^oS, IA_j^oS and IA_k^oS represent general expressions valid for any time-dependent situation of the type discussed above (see Equation 8). The reasons for this are given in a forthcoming report. The Equations for the unperturbed case have been given elsewhere, although in a slightly different form (9).

3. Activities During the Waiting Period

The kinetic equations describing the growth and decay of the radon (thoron) progeny after the sampling period are obtained by setting Q = 0 in Equations 9, 10 and 11, which then reduce to:

$$\frac{dA_i}{dt} = -\lambda_i A_i \qquad \text{Eq 24}$$

$$\frac{dA_j}{dt} = \lambda_j A_i - \lambda_j A_j \qquad \text{Eq 25}$$

$$\frac{dA_k}{dt} = \lambda_k A_j - \lambda_k A_k \qquad \text{Eq 26}$$

The activities during the waiting period (i.e., after sampling) are obtained upon integration of Equations 24, 25 and 26, taking into account the boundary conditions: $A_i(T_w = 0) = A_i(T_s)$, $A_j(T_w = 0) = A_j(T_s)$ and $A_k(T_w = 0) = A_k(T_s)$, where $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ are given by Equations 12, 14 and 16, respectively.

$$A_i(T_w) = A_i(T_s)e^{-\lambda_i T_w} \qquad \text{Eq 27}$$

$$A_j(T_w) = A_j(T_s)e^{-\lambda_j T_w} + A_i(T_s)\frac{\lambda_j}{\lambda_j-\lambda_i}\left(e^{-\lambda_i T_w} - e^{-\lambda_j T_w}\right) \qquad \text{Eq 28}$$

$$A_k(T_w) = A_k(T_s)e^{-\lambda_k T_w} + A_j(T_s)\frac{\lambda_k}{\lambda_k-\lambda_j}\left(e^{-\lambda_j T_w} - e^{-\lambda_k T_w}\right)$$

$$+ A_i(T_s)\lambda_j\lambda_k\left[\frac{e^{-\lambda_i T_w}}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{e^{-\lambda_j T_w}}{(\lambda_i-\lambda_j)(\lambda_k-\lambda_j)} + \frac{e^{-\lambda_k T_w}}{(\lambda_i-\lambda_k)(\lambda_j-\lambda_k)}\right] \qquad \text{Eq 29}$$

Equations 27 to 29 are general expressions applicable to any case,

i.e., unperturbed and perturbed. The only difference between cases is in $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ which assume different forms according to the case (9).

4. Integrated Activities During the Waiting Period

The integrated activities during any arbitrary waiting period $T_{w1} \rightarrow T_{w2}$ can be obtained by integration of Equations 27 to 29 over the desired waiting times. It is easy to show that:

$$IA_iW = A_i(T_s)\phi_i \qquad \text{Eq 30}$$

$$IA_jW = A_j(T_s)\phi_j + A_i(T_s)\frac{\lambda_j}{\lambda_j-\lambda_i}(\phi_i - \phi_j) \qquad \text{Eq 31}$$

$$IA_kW = A_k(T_s)\phi_k + A_j(T_s)\frac{\lambda_k}{\lambda_k-\lambda_j}(\phi_j - \phi_k) + A_i(T_s)\lambda_j\lambda_k\left[\frac{\phi_i}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{\phi_j}{(\lambda_i-\lambda_j)(\lambda_k-\lambda_j)} + \frac{\phi_k}{(\lambda_i-\lambda_k)(\lambda_j-\lambda_k)}\right] \qquad \text{Eq 32}$$

where,

$$\phi_i = (1/\lambda_i)(e^{-\lambda_i T_{w1}} - e^{-\lambda_i T_{w2}})$$

$$\phi_j = (1/\lambda_j)(e^{-\lambda_j T_{w1}} - e^{-\lambda_j T_{w2}})$$

$$\phi_k = (1/\lambda_k)(e^{-\lambda_k T_{w1}} - e^{-\lambda_k T_{w2}})$$

As with Equations 27 to 29, Equations 30 to 32 are general expressions applicable to any case.

LINEAR CASE

1. Activities During the Sampling Period

For the linear case, the activities during sampling are as follows:

$$A_i(T_s) = A_i^o(T_s) + BT_s - C(1 - e^{-\lambda_i T_s}) \qquad \text{Eq 33}$$

$$A_j(T_s) = A_j^o(T_s) + ET_s - F + \frac{\lambda_j C}{\lambda_j-\lambda_i}e^{-\lambda_i T_s} + D\,e^{-\lambda_j T_s} \qquad \text{Eq 34}$$

and,

$$A_k(T_s) = A_k^o(T_s)$$

$$+ HT_s - I + Je^{-\lambda_i T_s} + \frac{D\lambda_k}{\lambda_k-\lambda_j} e^{-\lambda_j T_s} + Le^{-\lambda_k T_s} \qquad \text{Eq 35}$$

where, $A_i^o(T_s)$, $A_j^o(T_s)$ and $A_k^o(T_s)$ are given by Equations 13, 15 and 17, and where:

$$B = \frac{QC_{oi}\eta}{\lambda_i} \ ,$$

$$C = \frac{QC_{oi}\eta}{\lambda_i^2} \ ,$$

$$D = \frac{QC_{oj}\eta+\lambda_j B}{\lambda_j^2} - \frac{C\lambda_i}{\lambda_j-\lambda_i} \ ,$$

$$E = \frac{QC_{oj}\eta + \lambda_j B}{\lambda_j} \ ,$$

$$F = C + \frac{QC_{oj}\eta + \lambda_j B}{\lambda_j^2} \ ,$$

$$H = \frac{QC_{ok}\eta + E\lambda_k}{\lambda_k} \ ,$$

$$I = F + \frac{QC_{ok}\eta + E\lambda_k}{\lambda_k^2} \ ,$$

$$J = \frac{\lambda_j\lambda_k C}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} \ , \text{ and}$$

$$L = I - J - \frac{D\lambda_k}{\lambda_k-\lambda_j} \ .$$

2. Integrated Activities During the Sampling Period

The integrated activities during the sampling period $T_{s1} \rightarrow T_{s2}$ are given as follows:

$$IA_iS = IA_i^oS + B\left(\frac{T_{s2}^2-T_{s1}^2}{2}\right) - C(T_{s2}-T_{s1}) + C\,G_i \qquad \text{Eq 36}$$

$$IA_jS = IA_j^oS + E\left(\frac{T_{s2}^2-T_{s1}^2}{2}\right) - F(T_{s2}-T_{s1}) + \frac{\lambda_j C}{\lambda_j-\lambda_i} G_i + D\,G_j \qquad \text{Eq 37}$$

$$IA_kS = IA_k^oS + H\left(\frac{T_{s2}^2 - T_{s1}^2}{2}\right) - I(T_{s2} - T_{s1}) + JG_i + \frac{D\lambda_k}{\lambda_k - \lambda_j} G_j + L\,G_k \qquad \text{Eq 38}$$

where IA_i^oS, IA_j^oS and IA_k^oS, and G_i, G_j and G_k, have already been defined in the text.

3. Activities During the Waiting Period

The activities during the waiting period are given by Equations 27, 28 and 29 where $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ are given by Equations 33, 34 and 35.

4. Integrated Activities During the Waiting Period

The integrated activities during the waiting period are given by Equations 30, 31 and 32 where, again, $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ are given by Equations 33, 34 and 35.

EXPONENTIAL CASE

1. Activities During the Sampling Period

In this case the activities during sampling are given by:

$$A_i(T_s) = \frac{QC_{oi}}{\delta + \lambda_i}\left(e^{\delta T_s} - e^{-\lambda_i T_s}\right) \qquad \text{Eq 39}$$

$$A_j(T_s) = M\,e^{\delta T_s} + (N-M)e^{-\lambda_j T_s} - N\,e^{-\lambda_i T_s} \qquad \text{Eq 40}$$

where,

$$M = \frac{QC_{oj}}{\delta + \lambda_j} + \frac{QC_{oi}\lambda_j}{(\delta + \lambda_i)(\delta + \lambda_j)}, \quad \text{and}$$

$$N = \frac{QC_{oi}\lambda_j}{(\delta + \lambda_i)(\lambda_j - \lambda_i)}$$

$$A_k(T_s) = Pe^{\delta T_s} - \frac{N\lambda_k}{\lambda_k - \lambda_i} e^{-\lambda_i T_s} + Re^{-\lambda_j T_s} + Se^{-\lambda_k T_s} \qquad \text{Eq 41}$$

where,

$$P = \frac{QC_{ok} + M\lambda_k}{\delta + \lambda_k}$$

$$R = \frac{\lambda_k(N-M)}{\lambda_k - \lambda_j}$$

$$S = \frac{N\lambda_k}{\lambda_k - \lambda_i} - (P+R)$$

2. Integrated Activities During the Sampling Period

Integration of Equations 39 to 41 gives:

$$IA_iS = \frac{QC_{oi}}{\delta+\lambda_i}(G_s - G_i) \qquad \text{Eq 42}$$

$$IA_jS = MG_s + (N-M)G_j - N\,G_i \qquad \text{Eq 43}$$

$$IA_kS = PG_s - \frac{N\lambda_k}{\lambda_k - \lambda_i} G_i + R\,G_j + S\,G_k \qquad \text{Eq 44}$$

where,

$$G_s = (1/\delta)(e^{\delta T_{s2}} - e^{\delta T_{s1}}) \quad ,$$

and G_i, G_j and G_k have already been defined above.

As with the other perturbation cases discussed above, the activities during the waiting period are given by Equations 27, 28 and 29, whereas the integrated activities during the waiting period are given by Equations 30, 31 and 32.

DISCUSSION AND CONCLUSIONS

The expressions for $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ given by Equations 9, 10 and 11, respectively, for the perturbation cases investigated here are quite complex algebraically. The above expressions exhibit some 'symmetries' which are not always fully apparent or reflected in the particular form given here, where symmetry of form has often been sacrificed for the sake of practicality.

As expected, the solutions for $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$ are proportional to C_{oi}, C_{oi} and C_{oj}, and C_{oi}, C_{oj} and C_{ok}, respectively. Furthermore, the following observations for the activities, for all perturbation cases, are worthwhile noticing. For any given perturbation case, the solutions to Equations 9 to 11:

a) contain terms proportional to the perturbation and its time derivative. It also contains an unperturbed term proportional to C_{om};

b) contain terms corresponding to the initial condition, i.e., for $t = 0$, multiplied by a decay factor of the form $e^{-\lambda_m t}$, where $m = i, j, k$;

c) contain exponential decay terms, as in item b), ranging from the first member of the chain to the particular radionuclide of interest.

The expressions for $A_i(T_w)$, $A_j(T_w)$ and $A_k(T_w)$ given by Equations 24, 25 and 26, respectively, only contain perturbation terms through $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$. Except for this, the expressions are identical in form to those given for the unperturbed case discussed elsewhere (9). The same arguments apply to the integrated activities after sampling, i.e., IA_iW, IA_jW and IA_kW.

The expressions for IA_iS, IA_jS and IA_kS are somewhat more complicated than those corresponding to their differential activities $A_i(T_s)$, $A_j(T_s)$ and $A_k(T_s)$, respectively. As expected, the integrated expressions contain time-dependent terms which are the result of the integration of time-dependent and time-independent terms in the solutions for the differential activities.

Broadly speaking, the solutions for the differential and integrated activities exhibit a similar, although somewhat modified, behaviour to that corresponding to the perturbation. Furthermore, because of the time-dependent nature of the perturbation, radioactive decay, and the different half-lives of the radionuclides involved, the differential activities for the different radionuclides are out of phase, as shown elsewhere (10).

Computer programs have been written to determine the differential and integrated activity deposited by active sampling on filter material during the sampling period and the waiting, i.e., after sampling, period. Calculations have been carried out under 'unperturbed' and time-dependent conditions. The data obtained for the radon and thoron progeny have been reported and discussed

elsewhere (10). The present work has practical applications to personal dosimetry and environmental monitoring (9).

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